SOME PROBLEMS IN NUCLEAR STRUCTURE STUDIED BY AN ELECTRON-ELECTRON COINCIDENCE SPECTROMETER

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ALMQVIST & WIKSELLS BOKTRYCKERI AB

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INAUGURAL DISSERTATION

BY

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FIL. LIC.

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To my Parents

Some problems in Nuclear structure studied by an elcctron-electron coincidence spectrometer

A radioactive nucleus often decays to an excited state in the daughter nucleus, and practically all excited states of nuclei are subject of decay to lower states with emission of electromagnetic radiation (gamma-rays) or internal conversion (where the transition to a lower state is then connected with the ejection of an atomic electron from a bound orbit). Other modes of decay may also occur depending on the particular state, e.g. beta decay and alpha decay, ... etc., provided the halflife of the excited state is long enough to allow for the transitions.

It was first pointed out by C.D. Ellis [1] and by Lise Meitner [2] in 1922 that the complex gamma-rays from some radioactive substances such as RaB exhibit simple additive rules,

$$h\nu_1 + h\nu_2 = h\nu_3$$

and hence that gamma-rays represent radiative transitions between quantized energy levels in the nucleus.

After the development of the quantum theory of radiation, it became possible to classify gamma-radiation into electric and magnetic multipole radiations of various multipole orders and to associate these with the difference in angular momentum and parity between the two states involved in the transition. The conservation of angular momentum and parity for the system of the nucleus and the electromagnetic radiation field imposes selection rules. These may be summarized as follows:

$$|J_i - J_f| \leqslant L \leqslant J_i + J_f$$

where L is the multipolarity of the radiation field.

J_i is the nuclear angular momentum (spin) of the initial state.

J_f is the nuclear angular momentum (spin) of the final state.

The character (electric or magnetic) of the radiation field is given by:

$$\Delta\pi = \frac{\pi_i}{\pi_f} = (-1)^L \text{ for electric multipole radiation.} \qquad (1)$$

$$= (-1)^{L-1} \text{ for magnetic multipole radiation.}$$

and $\Delta\pi = +1$ denotes no parity change.

$\Delta\pi = -1$ denotes parity change.

Where π_i is the parity of the initial state.

π_f is the parity of the final state.

An electric transition of multipole order L may then be represented as EL and the magnetic multipole as ML. When the selection rules allow for more than one multipole of a given character it is sufficient to consider only the lowest possible value of L. Higher order multipoles will have vanishing transition probabilities compared to the lowest order. However it may frequently happen that two multipoles of different characters are of comparable intensities (e.g. $M1 + E2$), such a transition is called mixed.

It should be observed that these quantum electrodynamic selection rules are independent of the intrinsic properties of the nucleus. There may also exist additional selection rules which depend on the details of the nuclear structure. Specific assumptions about nuclear structure i.e. nuclear models, generally impose special selection rules. Examples of such model dependent selection rules are l-forbidden $M1$ transitions in the single particle model and K-forbidden transitions in the collective model.

Internal conversion and gamma-ray emission may to a first approximation be considered as competing de-excitation processes. Therefore the total transition probability becomes:

$$\lambda_{\text{tot}} = \lambda_\gamma + \lambda_e \tag{1}$$

However under normal circumstances the conversion coefficient, i.e.

$$\alpha_x(EL, ML) = \frac{\lambda_{e_x}}{\lambda_\gamma} \tag{2}$$

depends only on the multipolarity and the character of the transition (EL or ML) and of the atomic shell from which the conversion electron originates, ($x = K, L_{\text{I}}, L_{\text{II}}, L_{\text{III}}$ ... etc.). The total conversion coefficient may then be defined as

$$\alpha_{\text{tot}} = \sum_x \alpha_x \tag{3}$$

For mixed transition we define the mixing ratio δ^2 as

$$\delta^2 = \lambda_{EL}/\lambda_{ML} \tag{4}$$

The conversion coefficient and the transition probability then become

$$\alpha_x(EL + ML) = \frac{\delta^2 \alpha_x(EL) + \alpha_x(ML)}{1 + \delta^2} \tag{5}$$

and

$$\lambda_{\text{tot}} = \lambda_{EL} + \lambda_{ML} + \lambda_e \tag{6}$$

From these formulae we derive the expressions for the partial transition rates

$$\lambda_{EL} = \lambda_{ML} \cdot \delta^2 \tag{7}$$

$$\lambda_{ML} = [t_{1/2} \cdot 1.44\,(1 + \alpha_{\text{tot}})\,(1 + \delta^2)]^{-1} \tag{8}$$

where $t_{1/2}$ is the halflife of the excited state.

The formulae for the partial transition rates depend on both α and δ^2. But it is in that case sufficient to determine only one of these quantities experimentally, since α and δ^2 are related by the formula (5) and α_{EL} and α_{ML} have been calculated theoretically by Rose [3] and also by Sliv and Band [4].

Already in 1921 Hahn [5] found experimentally the substance UZ obtained from the radioactive decay of UX_1 ($_{90}Th^{234}$) had the same atomic number and the same atomic weight as the already known substance UX_2 ($_{90}Pa^{234}$) which is also a daughter product of UX_1. The substance UZ is now known to be a metastable state in UX_2, with a halflife of 1.14 minutes. Since this first discovery, a great number of such metastable or isomeric excited states have been found. An isomeric state is conveniently defined as an excited state whose lifetime has been measured.

The first theoretical explanation of nuclear isomerism was given by von Weizsächer [6] in 1936. von Weizsächer showed that the transition probability from an excited state is strongly dependent on the multipolarity L. Each additional unit of L may increase the halflife by as much as 10^6. Therefore if in the decay of the excited nucleus the possible transitions according to the selection rules corresponding to high multipoles, the excited state may have a rather long halflife.

In 1950 the single particle model was developed. One of the most striking results of the single particle model was the explanation of the so-called "islands of isomerism", i.e. the existence of nuclei with long lived excited levels in certain regions in the periodic table.

Around 1950 the experimental techniques to study nuclear isomerism was also substantially improved by the introduction of the scintillation detector. As long as one was restricted to Geiger counters or similar detectors, it was impossible to measure halflives shorter than about one microsecond. Scintillation detectors, however, are about 1000 times faster. Therefore it became possible to extend the search for isomeric levels down to lifetimes of the order of a nanosecond (10^{-9} sec.). The so called fast-slow coincidence circuits and the delayed coincidence technique were developed in order to take full advantage of the fast response characteristics of scintillation detector for short lifetime measurements.

It was thought that the single particle model would be able to account for these short lifetimes as well.

In 1951 Blatt and Weisskopf [7] had calculated the transition probabilities on the assumption that the wavelength of the emitted radiation λ is much larger than the dimension R (the nuclear radius) of the radiating system (the nucleus) and that the radiating system consists of individual nucleons which contribute to the radiation due to the motion of their charges and magnetic moment only, i.e. no effects from exchange or mesic currents are considered.

The transition probabilities estimated by Blatt and Weisskopf are:

$$\lambda_{EL} = \frac{4 \cdot 4\,(L+1)}{L\,[(2L+1)!!]^2} \left(\frac{3}{L+3}\right)^2 \left(\frac{\hbar\omega}{197\ \text{MeV}}\right)^{2L+1} \times (R \text{ in } 10^{-13}\ \text{cm})^{2L}\, 10^{21}\ \text{sec}^{-1} \qquad (9)$$

$$\lambda_{ML} = \frac{1 \cdot 9\,(L+1)}{L\,[(2\,L+1)\,!\,!]^2}\left(\frac{3}{L+3}\right)^2\left(\frac{\hbar\omega}{197\ \mathrm{MeV}}\right)^{2L+1} \times (R \text{ in } 10^{-13}\ \mathrm{cm})^{2L-2}\, 10^{21}\ \mathrm{sec}^{-1} \qquad (10)$$

In the derivation of these expressions it was assumed that the nuclear wavefunction is always constant inside the nucleus and zero outside. The angular part of the wave function was not taken into account. Therefore the Blatt and Weisskopf formula does not contain the nuclear matrix element and no additional selection rules are imposed. It was found experimentally that the Blatt and Weisskopf formula explained generally reasonably well the observed halflives, but in certain cases large discrepancies were observed. It has become customary to compare the experimental halflives to Blatt and Weisskopf's estimates and to express the results in "units" of the Blatt and Weisskopf formula. Transitions which are faster than the values given by Blatt and Weisskopf are said to be favoured or enhanced and those slower than these are unfavoured or retarded.

Moszkowski [8] 1953 extended these calculations and took into consideration the harmonic oscillator wavefunction of the single particle model. Both the radial and the angular parts were considered. Moszkowski obtained:

$$\lambda_{\mathrm{sp}}(E\,L, \Delta J = L, \Delta l = L) = \frac{2\,(L+1)}{L\,[(2\,L+1)\,!\,!]^2}\,\frac{e^2}{\hbar}\left(\frac{\omega}{c}\right)^{2L+1} R^{2L}\, S\,(J_i, L, J_f)\,|\,m_{EL}|^2 \qquad (11)$$

$$\lambda_{\mathrm{sp}}(M\,L, \Delta J = L, \Delta l = L) = \frac{2\,(L+1)}{L\,[(2\,L+1)\,!\,!]^2}\,\frac{1}{\hbar}\left(\frac{\omega}{c}\right)^{2L+1} R^{2L-2}\left(\frac{e\hbar}{M\,c}\right)^2 S\,(J_i, L, J_f)\,|\,m_{ML}|^2 \qquad (12)$$

In addition to the selection rules for total angular momentum and parity change, a more stringent condition holds: $L \geqslant |L_i - L_f|$. In case of a single particle model, this forbids, for instance, $M1$ transitions between states $d\,3/2$ and $s\,1/2$ (i.e. $|\Delta j| = 1, |\Delta L| = 2$, no change of parity) which would be allowed in the general case. The statistical factor S is not very different from 1. The nuclear matrix elements would be expected to vary sufficiently from one nucleus to the other to overshadow any dependence on S. This is indeed so, for all transitions except for one important group, namely the $M4$ transitions.

Moszkowski's calculations did not differ much from those obtained by Blatt and Weisskopf and were in general in agreement with the experimental values. However, among the short lived isomer it was found that the transition probabilities for $E2$ and $M1$ deviates from the theoretical values obtained from the single particle model. The $E2$ multipole transitions are frequently enhanced by a considerable value over the single particle model estimates, while the $M1$ transitions, on the other hand are generally somewhat retarded. The enhancement of $E2$ was explained by Bohr and Mottelson [9] 1953 as due to the cooperative effects of a large number of nucleons. In fact the decay schemes contain levels which may be interpreted in terms of simple collective excitation of rotational or vibrational type.

In the vicinity of the closed shells however the single particle model is still con-

sidered to be the most suitable starting point. Theoretical studies of the nearly closed shell configurations, generally employ the single particle model wavefunction and extend the model by taking configurational mixing and (or) many particle configurations into consideration.

In order to study these recent developments and improvements of the nuclear shell model, a great deal of interest has been focused onto the very complex decay schemes. It is necessary for the study of such decay schemes to use a high resolution beta ray spectrometer in order to resolve the conversion lines, from which the transition energies are determined. A coincidence spectrometer may be used to establish the details of the decay schemes.

In the present thesis the decay of Tb^{160} has been studied by means of these experimental techniques. The decay was found to be fairly complex and nine excited levels were placed in the decay scheme. These levels are combined by sixteen nuclear transitions.

The remaining part of the present thesis deals with the studies of short lived excited states in regions near closed shell configurations. The main interest has been the study of the validity of the shell model. The partial transition rates λ_{M1} of the l-forbidden $M1$ transitions may be used as a sensitive check of the theory. It was found, in accordance with earlier observations, that the l-forbidden $M1$ transitions were much retarded, which seems to be a consequence of the l-selection rule discussed above. The matrix elements for these $M1$ transitions were calculated from Moszkowski's formula using the experimental values. They were found to be small but finite. It then becomes necessary to account for these values by extending the Moszkowski's theory replacing the shell model wavefunctions by more realistic nuclear wavefunction taking configurational mixing into account.

Sachs and Ross [10] 1951 suggested that the l-forbidden $M1$ transitions are due to exchange current effects.

Jensen and Mayer [11] 1952 have found that the presence of velocity, dependent interactions (e.g. spin orbit coupling, exchange forces ... etc.) affects the value of the transition probabilities. These interactions are not considered in Moszkowski's calculations of the transition probabilities. The velocity dependent forces, such as the spin orbit forces give rise to an anomalous $M1$ operator. This does not impose the l-retardation, which was found for the normal operator and the single particle shell model wavefunctions. Therefore transitions, which involve change of orbital angular momentum $|l_i - l_f| = l + 1$ can occur.

Recently Arima, Horie and Sano [12] 1957 have developed a theory based on configurational mixing which accounts very successfully for the static magnetic dipole moments or more precisely the deviation from the Schmidt-lines. The same authors have applied this theory to the l-forbidden $M1$ transitions. The agreement is less impressive but anyhow there is a considerable improvement over the single particle model.

The spectrometer with magnetic momentum selection in both channels has been found to be a powerful tool in studying the halflives of excited states. The delayed coincidence technique is the most general method to measure these transition rates. The ultimate limitations of the present technique are reviewed and it is concluded that certain sources of systematic errors can be eliminated if an electron-electron coincidence spectrometer is used, in combination with a fast coincidence unit.

The thesis contains the following papers:

I. E. Bashandy and J. Lindskog, A measurement of the lifetime of the first excited state in Tl^{200}, *Arkiv för Fysik 16*, 227 (1959).

II. J. Lindskog, E. Bashandy and T. R. Gerholm, Determination of a 7×10^{-11} sec. halflife for the first excited state in Tl^{201}, *Nuclear Physics 16*, 175 (1960).

III. E. Bashandy, Energy dependent instrumental time delays in millimicrosecond delayed coincidence experiments, *Nuclear Instruments and Methods 6*, 289 (1960).

IV. E. Bashandy, T. R. Gerholm and J. Lindskog, A delayed coincidence measurement of the 2.9×10^{-10} sec. halflife of the 279 keV first excited statein Tl^{203}, *Arkiv för Fysik 17*, 421 (1960).

V. G. Bäckström, J. Lindskog, O. Bergman, E. Bashandy and A. Bäcklin, Contributions to the decay of Tb^{160} to Dy^{160}, *Arkiv för Fysik 9*, 121 (1959).

The contents of the papers may be summarized as follows:

I. The lifetime of the 148 keV level in Tl^{200} has been measured with improved accuracy by the delayed coincidence method using the electron-electron coincidence spectrometer. A halflife of 7.3 ± 0.3 mμs has been found. The experimental transition probability for this $E2$ transition was compared with the transition probability based on the single particle model. The enhancementfactor P_{exp}/P_{theor} was of the same order of magnitude as the values obtained for other thallium isotopes.

II. A halflife of $(7 \pm 2) \times 10^{-11}$ sec. has been found for the first excited 330 keV state in Tl^{201}. The measurements have been performed with delayed coincidence technique. As the energy difference between the two transitions is only about 10 %, it was necessary to use a magnetic spectrometer to be able to resolve the lines. For this purpose the electron-electron coincidence spectrometer with a momentum resolution of 3 % and a transmission of 3 % was used. The measurements were performed according to the self comparison method. A new method to reduce the influence of energy dependent time delays has been developed. The $K/L_I + L_{II}$ and $L_I + L_{II}/L_{III}$ ratios for the 330 keV transition has been measured by means of an iron yoke double focusing spectrometer. From these measurements the mixing ratio $E2/M1$ was found to be 2.2 ± 0.4. The transition matrix element has been calculated and compared with those estimated by the single particle model and the theory by A. Arima *et al.*

III. The energy dependent time delays present in delayed coincidence measurements was studied experimentally. The most important factor among the various instru-

mental time delays contribute to the centroid shift was found to be due to the time lag between the absorption of the radiation of conversion electron in the crystal and the appearance of the first photoelectron. A large improvement obtained after careful adjustment of the detector system illustrates the importance of these factors in delayed coincidence experiments. These time delays are of the order of 10^{-10} sec. and may introduce serious systematic errors when submillimicrosecond lifetimes are to be measured by the centroid displacement method. By means of interpolation or extrapolation methods one can arrange the experimental conditions such that the energy dependent instrumental time delays do not contribute to the errors in the measurements. The ultimate limitation in delayed coincidence measurements is therefore under these condition set by the finite time resolution of the scintillation detectors and by the statistical accuracy.

IV. The measurements of the halflife of the 279 keV first excited level in Tl^{203} is reported. The experiment was carried out by the delayed coincidence technique. An electron-electron coincidence spectrometer was used for the energy resolution. A halflife of $(2.9 \pm 0.2) \times 10^{-10}$ sec. was found to be in good agreement with those obtained from delayed coincidence, nuclear resonance and coulomb excitation measurements. The reduced transition probability for the $E2$ transition and the matrix element for the $M1$ transition were calculated and compared with those obtained from single particle model.

V. The decay scheme of Dy^{160} has been re-investigated with high resolution and accuracy. The spectra of conversion electrons and photoelectrons have been measured. Four new transitions have been found. Their intensities as well as their multipolarities were determined. Coincidence measurements have been carried out between the different beta branches and the most prominent conversion lines. After these investigations it was possible to decide between two different decay chemes that have been proposed earlier by O. Nathan and by Clarc and Knowles. The possible existence of an odd parity vibrational band ($K = 0$) is discussed.

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